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(72) Inventors:
• **Majumdar, Biswaroop**
Delmar, New York 12054 (US)
• **Scobbo, James Joseph, Jr.**
Slingerlands, New York 12159 (US)

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(74) Representative: **Szary, Anne Catherine, Dr. et al**
London Patent Operation,
GE International, Inc.,
Essex House,
12-13 Essex Street
London WC2R 3AA (GB)

(71) Applicant: **GENERAL ELECTRIC COMPANY**
Schenectady, NY 12345 (US)

(54) **Method for preparing polyphenylene ether-polyamide resin blends having enhanced flow**

(57) The invention relates to a method of enhancing the melt flow characteristics of an impact modified composition of a compatibilized polyphenylene ether-polya-

mid resin blend. Compositions made by the method of the invention exhibit enhanced physical properties, especially enhanced melt flow and retention of properties.

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Description

[0001] The invention relates to a method of enhancing the melt flow characteristics of an impact modified composition of a compatibilized polyphenylene ether-polyamide resin blend.

[0002] The invention also relates to the compositions and articles formed out of the compositions made by the method of the invention.

[0003] Poly(phenylene ether) resins (referred to hereafter as "PPE") are commercially attractive materials because of their unique combination of physical, chemical, and electrical properties. Furthermore, the combination of these resins with polyamide resins into compatibilized blends results in additional overall properties such as chemical resistance, high strength, and high flow. Examples of such compatibilized blends can be found in U.S. Patents 4,315,086 (Ueno, et al); 4,659,760 (van der Meer); and 4,732,938 (Grant, et al). The properties of these blends can be further enhanced by the addition of various additives such as impact modifiers, flame retardants, light stabilizers, processing stabilizers, heat stabilizers, antioxidants and fillers.

[0004] The physical properties of PPE/polyamide blends make them attractive for a variety of end-use articles in the automotive market, especially for under hood and various exterior components. Many of these components are subjected to a variety of abuses such as impacts and as such require outstanding impact resistance and ductility. Moreover, many of these same articles are preferentially produced using conversion techniques such as injection molding. Some of the desirable applications, for example, connectors have very thin wall sections and therefore require resins that have very low viscosities in order to completely fill the molding tools. Conventional PPE/polyamide blends have inadequate flow properties at the processing temperatures that are needed to minimize the thermal degradation of the resins. Increasing the processing temperature to higher than these temperatures in order to reduce viscosity of the blends results in brittle parts and many surface imperfections in the final part, both of which are unacceptable.

[0005] It is therefore apparent that a need continues to exist for PPE/polyamide compositions that have improved melt flow yet retain the other attractive physical properties.

[0006] The needs discussed above have been generally satisfied by the discovery of an improved method for enhancing the melt flow characteristics of a PPE/polyamide composition, wherein the method comprises:

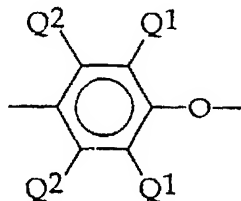
- (a) melt-mixing in a first step, from about 5% to about 50% by weight polyphenylene ether resin with from about 0.8% to about 1.2% by weight of a non-polymeric aliphatic polycarboxylic acid or derivative thereof, such as citric acid or a hydrate of citric acid and from about 2% to about 15% by weight of an elastomeric block copolymer, wherein the elastomeric block copolymer is a di-block copolymer, tri-block copolymer, or a mixture of a di-block copolymer and tri-block copolymer, wherein the copolymer comprises a polyarylene block and a saturated or unsaturated rubber block, with from about 5% to about 93% by weight of a polyamide resin, and
- (b) further melt mixing in a second step, from 0% to about 86% of a polyamide resin;

wherein the total weight of the polyamide resin is from about 35% to about 93% by weight based on the total weight of the composition.

[0007] In a preferred embodiment of the present invention, the PPE level is between about 15% and about 30% by weight, the citric acid level is between about 0.8% and about 1.0% by weight, the elastomeric block copolymer is present from about 6% to about 10% by weight, wherein elastomeric block copolymer contains a ratio of di-block to tri-block of about 1:3, the polyamide is a nylon 6,6 resin present at a level between about 60% and about 79% by weight, wherein between about 10% and 15% of the nylon 6,6 is added with the PPE, the citric acid, and the elastomeric block copolymer with the remainder of the nylon 6,6 added in a second step; wherein all weight percentages are based on the total weight of the composition.

[0008] The description which follows provides further details regarding this invention.

[0009] PPE, per se, are known polymers comprising a plurality of structural units of the formula (I):



(I)

wherein for each structural unit, each Q¹ is independently halogen, primary or secondary lower alkyl (e.g., alkyl con-

taining up to 7 carbon atoms), phenyl, haloalkyl, aminoalkyl, hydrocarbonoxy, or halohydrocarbonoxy wherein at least two carbon atoms separate the halogen and oxygen atoms; and each Q² is independently hydrogen, halogen, primary or secondary lower alkyl, phenyl, haloalkyl, hydrocarbonoxy or halohydrocarbonoxy as defined for Q¹. Preferably, each Q¹ is alkyl or phenyl, especially C₁₋₄ alkyl, and each Q² is hydrogen.

[0010] Both homopolymer and copolymer PPE are included. The preferred homopolymers are those containing 2,6-dimethyl-1,4-phenylene ether units. Suitable copolymers include random copolymers containing, for example, such units in combination with 2,3,6-trimethyl-1,4-phenylene ether units. Also included are PPE containing moieties prepared by grafting vinyl monomers or polymers such as polystyrenes, as well as coupled PPE in which coupling agents such as low molecular weight polycarbonates, quinones, heterocycles and formals undergo reaction in known manner with the hydroxy groups of two PPE chains to produce a higher molecular weight polymer, provided a substantial proportion of free OH groups remains.

[0011] The PPE generally has a number average molecular weight within the range of about 3,000-40,000 and a weight average molecular weight within the range of about 20,000-80,000, as determined by gel permeation chromatography. Its intrinsic viscosity is most often in the range of about 0.15-0.6 dl./g., as measured in chloroform at 25°C.

[0012] The PPE are typically prepared by the oxidative coupling of at least one monohydroxyaromatic compound such as 2,6-xenol or 2,3,6-trimethylphenol. Catalyst systems are generally employed for such coupling; they typically contain at least one heavy metal compound such as a copper, manganese or cobalt compound, usually in combination with various other materials.

[0013] Particularly useful PPE for many purposes are those which comprise molecules having at least one aminoalkyl-containing end group. The aminoalkyl radical is typically located in an ortho position to the hydroxy group. Products containing such end groups may be obtained by incorporating an appropriate primary or secondary monoamine such as di-n-butylamine or dimethylamine as one of the constituents of the oxidative coupling reaction mixture. Also frequently present are 4-hydroxybiphenyl end groups, typically obtained from reaction mixtures in which a by-product diphenoquinone is present, especially in a copper-halide-secondary or tertiary amine system. A substantial proportion of the polymer molecules, typically constituting as much as about 90% by weight of the polymer, may contain at least one of said aminoalkyl-containing and 4-hydroxybiphenyl end groups.

[0014] It will be apparent to those skilled in the art from the foregoing that the PPE contemplated for use in the present invention include all those presently known, irrespective of variations in structural units or ancillary chemical features.

[0015] The polyamide resins useful in the practice of the present invention are a generic family of resins known as nylons, characterized by the presence of an amide group (-C(O)NH-). Nylon-6 and nylon-6,6 are the generally preferred polyamides and are available from a variety of commercial sources. Other polyamides, however, such as nylon-4,6, nylon-12, nylon-6,10, nylon 6,9, nylon 6/6T and nylon 6,6/6T with triamine contents below about 0.5 weight percent as well as others, such as the amorphous nylons may be useful for particular PPE-polyamide applications. Mixtures of various polyamides as well as various polyamide copolymers are also useful. The most preferred polyamide for the blends of the present invention is polyamide-6,6.

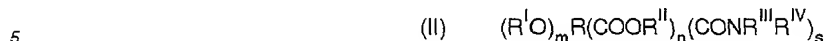
[0016] The polyamides can be obtained by a number of well known processes such as those described in U.S. Patent Nos. 2,071,250; 2,071,251; 2,130,523; 2,130,948; 2,241,322; 2,312,966; and 2,512,606. Nylon-6, for example, is a polymerization product of caprolactam. Nylon-6,6 is a condensation product of adipic acid and 1,6-diaminohexane. Likewise, nylon 4,6 is a condensation product between adipic acid and 1,4-diaminobutane. Besides adipic acid, other useful diacids for the preparation of nylons include azelaic acid, sebacic acid, dodecane diacid, as well terephthalic and isophthalic acids. Other useful diamines include m-xylene diamine, di-(4-aminophenyl)methane, di-(4-aminocyclohexyl)methane; 2,2-di-(4-aminophenyl)propane, and 2,2-di-(4-aminocyclohexyl)propane. Copolymers of caprolactam with diacids and diamines are also useful.

[0017] Polyamides having viscosity number of about 90 to about 350 ml/g, preferably between about 110 and about 240 ml/g as measured in a 0.5 weight percent solution in 96 weight percent sulphuric acid in accordance with ISO 307 is preferred.

[0018] In the method of the present invention, a compatibilizing agent should be employed in the preparation of the composition. The two-fold purpose for using a compatibilizing agent is to improve, in general, the physical properties of the PPE-polyamide resin blend, as well as to enable the use of a greater proportion of the polyamide component. When used herein, the expression "compatibilizing agent" refers to those polyfunctional compounds which interact with either the PPE, the polyamide resin, or both. This interaction may be chemical (e.g. grafting) or physical (e.g. affecting the surface characteristics of the dispersed phases). In either instance the resulting PPE-polyamide composition appears to exhibit improved compatibility, particularly as evidenced by enhanced impact strength, mold knit line strength and/or elongation. As used herein, the expression "compatibilized polyphenylene ether-polyamide base resin" refers to those compositions which have been physically or chemically compatibilized with an agent as discussed above, as well as those compositions which are physically compatible without such agents, as taught in U.S. Pat. No. 3,379,792.

[0019] In the practice of the present invention, non-polymeric aliphatic polycarboxylic acids are used as the compatibilizing agent. Included in the group of non-polymeric aliphatic polycarboxylic acids species, also known as compati-

bilizers or functionalizers, are, for example, the aliphatic polycarboxylic acids, and acid esters represented by the formula (II)



wherein R is a linear or branched chain, saturated aliphatic hydrocarbon of from 2 to 20, preferably 2 to 10, carbon atoms; each R^I is selected from the group consisting of hydrogen or an alkyl, aryl, acyl, or carbonyl dioxy group of 1 to 10, preferably 1 to 6, most preferably 1 to 4, carbon atoms, with hydrogen being especially preferred; each R^{II} is independently selected from the group consisting of hydrogen or an alkyl or aryl group of from 1 to 20 carbon atoms preferably from 1 to 10 carbon atoms; each R^{III} and R^{IV} is independently selected from the group consisting essentially of hydrogen or an alkyl or aryl group of from 1 to 10, preferably from 1 to 6, most preferably 1 to 4, carbon atoms; m is equal to 1 and (n+s) is greater than or equal to 2, preferably equal to 2 or 3, and n and s are each greater than or equal to zero; and wherein (OR^I) is alpha or beta to a carbonyl group and at least two carbonyl groups are separated by 2 to 6 carbon atoms. Obviously, R^I , R^{II} , R^{III} , and R^{IV} cannot be aryl when the respective substituent has less than 6 carbon atoms.

[0020] Illustrative of suitable polycarboxylic acids are citric acid, malic acid, and agaricic acid; including the various commercial forms thereof, such as, for example, the anhydrous and hydrated acids. Illustrative acid esters useful herein include, for example, acetyl citrate and mono- and/or di- stearyl citrates and the like. Suitable acid amides useful herein include, for example, N,N'-diethyl citric acid amide; N,N'-dipropyl citric acid amide; N-phenyl citric acid amide; N-dodecyl citric acid amide; N,N'-didodecyl citric acid amide and N-dodecyl malic acid amide. Derivatives of the foregoing polycarboxylic acids are also suitable for use in the practice of the present invention. Examples of suitable functionalizing compounds can be found in U.S. Patent Nos. 4,315,086, 4,755,566, 4,873,286 and 5,000,897, all of which are incorporated herein by reference.

[0021] The amount of the above mentioned non-polymeric aliphatic polycarboxylic acids that is required to appropriately functionalize the PPE is that which is sufficient to improve the compatibility between the various components in the composition and improve the flow characteristics of the composition without loss of desirable properties such as, for example, impact properties, tensile properties, surface appearance, and the like. As previously discussed, indications of improved compatibility include resistance to lamination, improved physical properties such as increased tensile and impact properties and a stabilized morphology between the blend component phases under static or low shear conditions.

[0022] It is thought that reactions can occur between some of the components of the composition, for example, between the PPE and the non-polymeric aliphatic polycarboxylic acids, the non-polymeric aliphatic polycarboxylic acids and the polyamide resin, or between the PPE, non-polymeric aliphatic polycarboxylic acid, and the polyamide resin. These reactions are thought to lead to various copolymers between the components of the blend. In this manner, as the non-polymeric aliphatic polycarboxylic acid is added to a PPE/polyamide composition, the viscosity of the composition increases due to the formation of the copolymer structures. There appears to be a minimum threshold level of non-polymeric aliphatic polycarboxylic acid that is required to achieve the desired compatibility between the PPE and the polyamide. An increase in the level of non-polymeric aliphatic polycarboxylic acid above the minimum has little effect on enhancing the compatibility but unexpectedly has been discovered to have a significant effect on improving the melt flow characteristics of the blend.

[0023] In the practice of the present invention, an effective amount of the above mentioned non-polymeric aliphatic polycarboxylic acids, based on the total weight of the composition, is from about 0.8% to about 1.2% by weight, preferably from about 0.8% to about 1.0% by weight. Amounts in excess of about 1.2% by weight afford increased flow characteristics but at the expense of low temperature impact strength and the color of the composition darkens considerably. The amount of PPE in the present invention is between about 5% and about 50% by weight, preferably between about 10% and about 35% by weight, and most preferably between about 15% and 30% by weight, wherein the weight percentages are based on the entire weight of the composition. The amount of the polyamide is between about 35% and about 93% by weight, preferably between about 50% and about 88% by weight, and most preferably between about 55% and 83% by weight, wherein the weight percentages are based on the entire weight of the composition.

[0024] The method according to the invention incorporates in the composition one or more additional impact modifiers. All impact modifiers as generally used for compositions comprising a polyphenylene ether, a polyamide or a combination of a polyphenylene ether and a polyamide can be used. Particularly suitable are the so called block copolymers, for example, A-B-A triblock copolymers and A-B diblock copolymers. The A-B and A-B-A type block copolymer rubber additives which may be used are thermoplastic rubbers comprised of one or two alkenyl aromatic blocks which are typically styrene blocks and a rubber block, e.g., a butadiene block which may be partially hydrogenated. Mixtures of these triblock copolymers and diblock copolymers are especially useful in the present compositions.

[0025] Suitable A-B and A-B-A type block copolymers are disclosed in, for example, U.S. Patent Nos. 3,078,254, 3,402,159, 3,297,793, 3,265,765, and 3,594,452 and U.K. Patent 1,264,741, all incorporated herein by reference. Examples of typical species of A-B and A-B-A block copolymers include polystyrene-polybutadiene (SBR), polystyrene-poly(ethylene-propylene), polystyrene-polyisoprene, poly(α -methylstyrene)-polybutadiene, polystyrene-polybutadiene-polystyrene (SBR), polystyrene-poly(ethylene-propylene)-polystyrene, polystyrene-polyisoprene-polystyrene and poly(α -methylstyrene)-polybutadiene-poly(α -methylstyrene), as well as the selectively hydrogenated versions thereof. Mixtures of the aforementioned block copolymers are also useful. Such A-B and A-B-A block copolymers are available commercially from a number of sources, including Phillips Petroleum under the trademark SOLPRENE, Shell Chemical Co., under the trademark KRATON, and Kuraray under the trademark SEPTON.

[0026] A useful amount of impact modifier is between about 2% and about 15% by weight, preferably between about 4% and about 12% by weight, wherein the weight percentages are based on the entire weight of the composition. When both di-block and tri-block copolymers are utilized, the weight ratio of di-block to tri-block is between about 5:1 and about 1:6, preferably between about 4:1 and about 1:5, and most preferably between about 2:3 and about 1:4. In an especially preferred embodiment, the weight ratio of di-block to tri-block is between about 1:7 and about 3:5.

[0027] Compositions of the present invention can also include effective amounts of at least one additive selected from the group consisting of antioxidants, flame retardants, drip retardants, dyes, pigments, colorants, stabilizers, anti-static agents, plasticizers and lubricants. These additives are known in the art, as are their effective levels and methods of incorporation. Effective amounts of the additives vary widely, but they are usually present in an amount up to about 50% or more by weight, based on the weight of the entire composition. Especially preferred additives include hindered phenols, thio compounds and amides derived from various fatty acids. The preferred amounts of these additives generally ranges up to about 2% total combined weight based on the total weight of the composition.

[0028] The preparation of the compositions of the present invention is normally achieved by merely blending the ingredients under conditions for the formation of an intimate blend. Such conditions often include mixing in single or twin screw type extruders or similar mixing devices which can apply a shear to the components.

[0029] All of the ingredients may be added initially to the processing system, or else certain additives may be precompounded with one or more of the primary components, preferably the PPE, impact modifier and the polyamide. It appears that certain properties, such as impact strength and elongation, are sometimes enhanced by initially precompounding the PPE and impact modifier, optionally with any other ingredients, prior to compounding with the polyamide resin, however, these improvements are done at the expense of increasing the viscosity of the compatibilized composition. In the present invention, it is preferable that at least 5% by weight, preferably at least 8% by weight, and most preferably, at least 10% by weight polyamide be added with the PPE and non-polymeric carboxylic acid. The remaining portion of the polyamide is fed through a port downstream. In this manner, the viscosity of the compatibilized composition is reduced without significant reduction in other key physical properties. While separate extruders may be used in the processing, these compositions are preferably prepared by using a single extruder having multiple feed ports along its length to accommodate the addition of the various components. It is often advantageous to apply a vacuum to the melt through at least one or more vent ports in the extruder to remove volatile impurities in the composition. Those of ordinary skill in the art will be able to adjust blending times and temperatures, as well as component addition, without undue additional experimentation.

[0030] It should be clear that compositions and articles made from the compositions made by the method of this disclosure are within the scope of the invention.

[0031] All patents and patent applications cited are incorporated herein by reference.

[0032] The invention will be further illustrated by the following examples.

EXPERIMENTAL

[0033] In the examples the following materials have been used:

PPE: a poly(2,6-dimethyl-1,4-phenylene ether) with an intrinsic viscosity of 45 ml/g as measured in toluene at 25°C. and a concentration of 0.6 gram per 100 ml;

PA-6,6: a polyamide-6,6 with a viscosity of 35 ml/g according to ASTM D789 in a solution of 8.4% by weight of nylon in 90% Formic Acid;

CAH: citric acid monohydrate;

SEBS: polystyrene-poly(ethylene-butylene)-polystyrene block copolymer;

SEP: polystyrene-poly(ethylene-propylene) block copolymer;

[0034] The ingredients were compounded in the weight ratios as indicated in the following table in a twin-screw extruder with temperature settings over the length of the extruder between about 280 and about 310° C. The screw speed was 300 rpm, the throughput 10 kilograms per hour. All ingredients with exception of the polyamide were fed at

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the throat of the extruder; the polyamide was split-fed partially into the throat of the extruder with the remainder fed downstream about halfway the length of the extruder. The strands coming from the extruder were pelletized in a laboratory equipment and dried for about 3 hours at about 110°C. The dried pellets were injection molded into standard ASTM test specimens for measurement of physical properties.

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55 50 45 40 35 30 25 20 15 10 5

Sample:	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
PPE	21	21	21	21	21	21	21	21	21	21	21	21	21	21	21
CAH	0.5	0.6	0.7	0.75	0.8	0.85	0.9	0.95	1.0	1.05	1.1	1.15	1.2	1.3	1.4
SEP	6	6	6	6	6	6	6	6	6	6	6	6	6	6	6
SEBS	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
PA-6,6 ¹	13	13	13	13	13	13	13	13	13	13	13	13	13	13	13
PA-6,6 ²	54	54	54	54	54	54	54	54	54	54	54	54	54	54	54
Properties															
Viscosity ³	94	97	90	93	87	89	87	86	87	85	73	74	76	69	66
% decrease	0	+3	-3	-1	-5	-5	-7	8	-7	-11	-22	-21	-19	-27	-30
NI ⁴	3.6	3.7	3.7	3.6	3.6	4.0	3.5	3.3	3.6	3.6	3.6	3.8	3.7	3.7	3.5
Dyn. ⁵	21	26	36	27	35	35	28	29	31	32	34	35	34	34	24
TS ⁶	8.7	8.6	8.7	8.7	8.7	8.5	8.7	8.9	8.6	8.7	8.8	8.6	8.5	8.6	8.6
TE ⁷	49	50	47	54	45	55	48	38	51	45	46	50	50	48	50

¹ denotes amount of PA-6,6 added with the PPE;

² denotes amount of PA-6,6 added downstream;

³ viscosity is reported in Pa·Sec and was measured at $1500s^{-1}$ at 280°C;

⁴ NI stands for notched Izod impact reported in ft-lbs/in and tested according to D256;

⁵ Dyn. stands for Dynatup impact strength (energy to fracture falling dart test) reported in ft-lbs and measured according to D3783;

⁶ TS stands for tensile strength at yield reported in Kpsi and measured according to ASTM D638;

⁷ TE stands for tensile elongation at break reported in percent and measured according to ASTM D638. All compositions contain the following additives: 0.5 Seenox 412S, 0.3 potassium iodide, 0.5 Irganox 1010, 0.3 pentaerythritol tetraacetate, 0.05 copper (I) iodide, 0.5 of a 20% by weight carbon black concentrate in nylon-6.

[0035] As can be seen from the results of the table, the compositions containing at least 0.8% by weight citric acid unexpectedly had a 5% or greater decrease in their viscosity as measured at 1500s^{-1} as compared to a composition containing 0.5% by weight citric acid while substantially maintaining other physical properties. Moreover, compositions containing at least 1.05% by weight citric acid unexpectedly had a 10% or greater decrease in their viscosity as measured at 1500s^{-1} . A previously mentioned, compositions containing more than 1.2% by weight citric acid were darker in color and had diminished low temperature impact properties.

Claims

1. A method for enhancing the melt flow characteristics of a polyphenylene ether resin/polyamide resin composition, wherein the method comprises:

(a) melt-mixing in a first step, from about 5% to about 50% by weight polyphenylene ether resin with from about 0.8% to about 1.2% by weight of a non-polymeric aliphatic polycarboxylic acid or derivative thereof and from about 2% to about 15% by weight of an elastomeric block copolymer, wherein the elastomeric block copolymer is a di-block copolymer, tri-block copolymer, or a mixture of a di-block copolymer and tri-block copolymer, wherein the copolymer comprises a polyarylene block and a saturated or unsaturated rubber block, optionally with from about 5% to about 93% of a polyamide resin, and

(b) further melt mixing in a second step, from about 0% to about 86% by weight of a polyamide resin;

wherein the total amount of polyamide resin is from about 35% to about 93% by weight; and wherein all weights are based upon the entire weight of the composition.

2. The method of claim 1, wherein the method is done in an extruder and wherein the first step and the second step are done sequentially in the same extruder.

3. The method of claim 1, wherein the non-polymeric aliphatic polycarboxylic acid or derivative thereof is citric acid or a hydrate of citric acid present in an amount from about 0.8% to about 1.0% by weight.

4. The method of claim 1, wherein the amount of polyphenylene ether resin is from about 10% to about 35% by weight and wherein the total amount of polyamide resin is from about 50% to about 88% by weight.

5. The method of claim 1, wherein the amount of polyphenylene ether resin is from about 15% to about 30% by weight and wherein the total amount of polyamide resin is from about 55% to about 83% by weight.

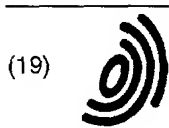
6. The method of claim 4, wherein the amount of elastomeric block copolymer is from about 4% by weight to about 12% by weight based on the weight of the entire composition.

7. The method of claim 5, wherein the amount of elastomeric block copolymer is from about 4% by weight to about 12% by weight based on the weight of the entire composition.

8. The method of claim 1, wherein the elastomeric block copolymer is a mixture of a di-block copolymer and a tri-block copolymer.

9. The method of claim 1, further comprising melt mixing in the first step, at least one additive selected from the group consisting of reinforcing fibers, fillers, conductive carbon black, carbon fibers and carbon fibrils, stabilizers, dyes, pigments, polyolefins, flame retardants, and mixtures thereof.

10. A composition made from the method of any preceding claim.



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(71) Applicant: **GENERAL ELECTRIC COMPANY**
Schenectady, NY 12345 (US)

(72) Inventors:
• **Majumdar, Biswaroop**
Delmar, New York 12054 (US)
• **Scobbo, James Joseph, Jr.**
Slingerlands, New York 12159 (US)

(74) Representative: **Szary, Anne Catherine, Dr. et al**
London Patent Operation,
GE International, Inc.,
Essex House,
12-13 Essex Street
London WC2R 3AA (GB)

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EUROPEAN SEARCH REPORT

Application Number
EP 99 30 0811

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
X	US 5 475 049 A (OHTOMO TAKASHI ET AL) 12 December 1995 (1995-12-12)	1-4,6,9,10	C08J3/00 C08K5/092
Y	* column 10, line 18-44; claims 1,16 * * column 12, line 29-38; tables 2,3 *	5,7,8	C08L71/12 C08L77/00
X	EP 0 395 994 A (GEN ELECTRIC) 7 November 1990 (1990-11-07)	1-7,9,10	
Y	* page 3, line 47,48; claims 1,9-15,25 * * page 4, line 57 * * page 5, line 45 - page 6, line 5 * * page 6, line 34-41; example 1 *	8	
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